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⑮ 発明の名称 汚水の浄化方法及び装置

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## 明細書

## 1 発明の名称

汚水の浄化方法及び装置

## 2 特許請求の範囲

1. 金属鉄を混入した土壌或いは充填材の屑に汚水を供給して汚水中の酸素を消費することにより土壌或いは充填材の屑を嫌気的雰囲気に保ち、脱窒菌の活性を向上させて脱窒を効果的に行わせることを特徴とする汚水の浄化方法。
2. 汚水を透水・好気的な土壌或いは充填材の屑に通水して汚水中のアンモニア態窒素を硝化させたのち、金属鉄を混入した土壌或いは充填材の屑に浸透させて脱窒させることを特徴とする汚水の浄化方法。
3. 金属鉄とともに、炭素源を混入するものである請求項1又は請求項2記載の汚水の浄化方法。

4. 汚水供給源の下方に、透水・好気的な土壌或いは充填材の屑又はブロックと、金属鉄を混入した土壌或いは充填材の屑又はブロックとを組み合わせて配置したことを特徴とする汚水の土壌式浄化装置。

## 3 発明の詳細な説明

## 【産業上の利用分野】

本発明は、汚水の処理方法、特に生活排水や屎尿処理水、下水等の汚水中に含まれる窒素分や磷分を土壌を利用して高度に処理する新規な方法及び装置に関する。

## 【従来の技術】

従来実施されている土壌浄化法の問題点、及び土壌に要求される性質は、以下の3点に要約される。

- ① 透水性が十分高く、目詰りが起きにくいこと。
- ② 活性アルミニウムや活性鉄等の磷酸を吸着できる化合物の含量が高いこと。

⑤ アンモニア態窒素の硝化に必要な好気的な土壤と、微生物による脱窒作用に必要な炭素源の供給が可能でしかも相対的に嫌気的な土壤層が共存し、汚水は両層を十分な速さで通過し、且つ両層に十分接触浸透できる構造になっていること。

この、相矛盾する条件を共に満足させうるものとして、本発明者は透水性及び透水性に優れた土壤層（砂、マサ土、ゼオライト粒等の層：以下「透水・好気性土壤層」と言う）と、透水性は劣るが活性アルミニウムや活性鉄及び炭素源に富む土壤層（黒ボク、赤土等の層：以下「難透水・嫌気性土壤層」と言う）を組み合わせた、多段土壤層法とでも言うべき理想的な土壤浄化方法及び装置を開発した（特願昭60-52729、特開昭61-212386）。

更に、第4図に示すように、難透水・嫌気性土壤層16を透水性のある容器や袋体13に詰めた一種の土壤ブロック17として難透水・嫌気性土壤層を形成することによって、施工性の問題を解決

した。また、これらの容器や袋体の素材として、木材やジュート等の炭素率（C/N比）の高いものを用いることによって、脱窒能力の向上を確保した（特願昭61-10730）。

#### 【発明が解決しようとする課題】

ところが、上記した従来の多段土壤層法による装置については、特に難透水・嫌気性土壤（土壤ブロック）にいくつかの問題が存在する。即ち、透水・好気性土壤はアンモニア態窒素の硝化に必要な条件を十分満たしており、また高負荷に耐える良好な透水性を示し何ら問題はない。

これに対し、難透水・嫌気性土壤は微生物による脱窒作用に必要な炭素源の供給は十分であったが、嫌気的な土壤層として考慮した場合、脱窒能力の安定化及び微生物（脱窒菌）の活性の高さの点ではまだ改善の余地が認められた。即ち、空素浄化能は脱窒菌の脱窒反応速度によって規定されるが、汚水は難透水・嫌気性土壤中をあまり通過せず主としてブロックの周縁部を通過する。その

ため、流入負荷量（L/m<sup>2</sup>・日）を増やすと周縁部での流速が速くなって浄化能が低下する。また、流入負荷が一定の場合は多段土壤層の厚みと浄化能は比例するが、容量負荷量（L/m<sup>2</sup>・日）を一定とし土壤層の厚みを厚く（例えば2倍）して流入負荷量を増やす（例えば2倍）と、浄化率は悪化する傾向にある。従って、装置の汚水処理可能容量（L/m<sup>2</sup>・日）を増やすには面積を大きくする必要がある。そのため、施工場所が限定されるし、高コストになり実施化に大きな障害となる。

#### 【課題を解決するための手段】

本発明は上記に描みなされたもので、脱窒作用に重要な影響を及ぼす難透水・嫌気性土壤に代わって、透水性に優れた土壤や充填材に金属鉄を混合した易透水・嫌気性の改良土壤（以下「易透水・嫌気性土壤」と言う）を使用することにより、処理能力の増大とともに、脱窒菌の活性を向上させて脱窒能力を飛躍的に向上できるようにするものである。

即ち、還元鉄等の金属鉄が空気を含んだ水と接触した場合、中性域では空気中の酸素による酸化作用により微量の鉄イオンが溶出する。この現象を利用して、汚水中の酸素を消費することにより土壤層を嫌気的雰囲気に保ち、脱窒菌の活性を向上させる。更に、嫌気性土壤部分の透水性を良くして、万遍なく汚水が浸透して脱窒作用を良好に行わせんとするものである。

金属鉄は、必ずしも純鉄に限らない。また、反応性の点では還元鉄が好ましいが、必ずしもこれに限らない。金属鉄の形状は、取り扱い易さや溶性を考慮すると、粒状のものが好ましい。その大きさは、通常5～20メッシュ程度のものを用いる。

金属鉄の使用割合は、金属鉄の純度や粒の大きさ、原水（汚水）中の空素濃度や溶存酸素量、処理水量等などを基に計算或いは実験によって決定される。鉄粒の場合、通常2～10重量%特に4～6%程度が好ましい。これより少ないと、脱窒効率が落ちるし、多すぎると鉄イオンの溶出の問

題が生じる。

尚、金属鉄が溶解して生じる鉄イオンは、硝酸イオンと結合して沈殿するので、硝の除去にも優れた効果を示す。

易透水・嫌気性土壌層を構成する土壌としては、砂、マサ土の他、砂丘未熟土、粗粒火山灰土、粗粒褐色森林土等透水性の優れた土壌が用いられる。また、土壌の替わりに用いられる充填材としては、ゼオライト粒やバーライト、バーミキュライト等の天然或いは人工の粒状鉱物の他、プラスチック粉碎品等も用いられる。これらの土壌や充填材中の炭素含量が少ない場合は、炭素源として、ジュート、稻藁、木の葉、その他の動植物体、余剰活性汚泥等炭素率(C/N比)の高い物質を混入しておいてもよい。

易透水・嫌気性土壌は、そのまま装置内に充填してもよいが、透水性のある容器や袋体に詰めた一種の土壌ブロックとすると、取り扱いが極めて簡単になる。また、金属鉄と土壌との比重の違いによる装置全体としての金属鉄の偏在も防止され

るし、透水・好気性土壌との使用割合も設計通りにできる等の利点も生じる。また、これらの容器や袋体の素材として、木材やジュート等の炭素率(C/N比)の高いものを用いると、脱空能力の向上が図れる。

一方、前記易透水・嫌気性土壌層或いはブロックの間に充填される透水・好気性土壌としては、砂やマサ土等前記易透水・嫌気性土壌と同じものの他に、同じくゼオライト粒その他の充填材も用いられる。

この透水・好気性土壌の主要な役割は、汚水を易透水・嫌気性土壌の層やブロックになるべく効率的に接触、拡散、浸透できるようにするとともに、装置の目詰りを防止して速やかに透水させることである。また、この透水・好気性土壌を中心として、SS分、BOD及びCOD分その他の有機物の好気的分解や硝化、脱臭等が行なわれる。従って、この土壌には通気性及び透水性が大きいこと(例えば、飽和透水性係数が $10^{-2} \sim 10^{-3} \text{ cm/s}$ よりも大)が要求される。場合によっては、

砂や砾、適当な大きさの木の枝や人工芝等、透水性を促進させるもを混入してもよい。

透水・好気性土壌として、ゼオライト粒を用いた場合には、ゼオライトはアンモニウムイオン保持能が大きく、吸着されたアンモニウムイオンは硝化菌の作用を受けて硝酸態窒素に変化し、ゼオライト粒から離脱する。そして、再びアンモニウムイオンが吸着されるという過程が繰り返される。こうした挙動は装置内における窒素の滞留時間を長くする効果を持ち、窒素除去に有利に働くことになる。更に、ゼオライトの大きなECは硝化に伴う汚水のpH低下に対する緩衝作用を持っており、装置内における微生物活動を保護するなど好ましい作用を行なうものである。

#### 【作用】

しかして、第1図で模式的に示すように、汚水供給源としての汚水撒水管1から供給された汚水(原水)(A)は、マサ土等からなる被覆土壌層2中で、土壤生物の消化分解作用や土の吸着や滤

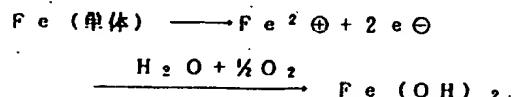
過作用によりSS分やBOD及びCOD成分その他の有機物の好気的分解や除去作用を受ける。またアンモニア態窒素も硝化細菌等の作用で硝化されて、処理水(B)となる。

この処理水(B)の一部は被覆土壌層2の表面から蒸散するが、大部分はトレンチ3下方の浄化層4に重力的に浸透流下する。浄化層4は、透水・好気性土壌5の層と易透水・嫌気性土壌6の層を複数層(図では2層)積層したものである。浄化層4としては、易透水・嫌気性土壌6をジュート製袋体等に充填した易透水・嫌気性土壌ブロックを、上下と前後左右を間隙をあけて配置し、その間際に透水・好気性土壌5を充填したものでもよい。

透水・好気性土壌5中に浸透した処理水(B)はより酸化的条件下に置かれ、被覆土壌層2と同様に有機物の好気的分解や硝化作用を受ける。尚、ゼオライト粒を用いている場合は、ここでアンモニア態窒素の固定や硝化も行なわれる。

次いで、処理水(B)は易透水・嫌気性土壌6

中に浸透し、その中に含まれる還元剤例えば鉄粒に接触して、次の反応を生じる。



そのため、処理水 (B) や易透水・嫌気性土壌 6 中の酸素を多量に消費する。この作用によって易透水・嫌気性土壌 6 の層全体が常に嫌気性に保たれ、脱空菌の活性が向上する。従って、処理水 (B) 中の  $\text{NO}_3^-$  や  $\text{NO}_2^- - \text{N}$  (硝酸態窒素) は、該土壌 6 の層中を浸透流下する時に脱空菌によって  $\text{N}_2$  や  $\text{N}_2\text{O}$  に変化し、効率的に脱空される。またこの過程で、処理水 (B) 中の磷酸 (正及びボリ) は易透水・嫌気性土壌中の鉄イオン  $\text{Fe}^{2+}$  と反応して磷酸鉄の沈殿となり、土壌 6 の層中に吸着固定される。

かくして、SS 分、BOD 及び COD 成分その他の有機物に加えて窒素分や磷が大幅に除去された浄化水 (C) が、排水層 7 に集められ、排水管 8 を通って装置外に排出される。

部  $3\text{cm} \times 2.5\text{cm} \times 10\text{cm}$  サイズのジュート製袋体 13 に充填 (200 g) して易透水・嫌気性土壌ブロック 14 とし、これを、上下と左右に 5 mm の間隔をおいて並べた。各層の土壌ブロック 14 は、処理水 (B) が十分に接触浸透できるように 2.5 cm ずつずらして配置した。使用した土壌ブロック 14 は 77 個で 9 段積みした。

このジュート製袋体 13 は、単に易透水・嫌気性土壌を充填するユニットを構成しているだけでなく、それ自体好気的土壌と嫌気的土壌の界面に存在する網状体であり、両層の接触界面であらゆる方向に水の浸透・移動を可能にしている。またジュート製袋体 13 は、炭素率 (C/N 比) が極めて高い (通常 5.0 以上) ので脱空菌の炭素源ともなり、装置の脱空活性を高める働きもする。尚、前記汚水浄化装置 9 の構造や土壌ブロック 14 の素材形状はあくまでも一例であり、本発明はこれらに限定されるものではない。

しかし、この汚水浄化装置 9 に、原水 (A) として人工汚水 ( $\text{NO}_3^- - \text{N} 4.0 \text{mg/l} + \text{PO}_4^{3-}$ )

### 【実施例】

次に、本発明を図面に示す実施例に基づいて詳細に説明する。

第 2 図は、本発明に係る実験室規模の汚水浄化装置の一例を示す。この汚水浄化装置 9 は、巾 10 cm、長さ 45 cm、深さ 45 cm の内法寸法のアクリル製の槽 10 の中に各土壌を納めたものである。即ち、上部から汚水吸水管 1 を配置した被覆土壌層 2、中央部は浄化層 4、下部は排水管 8 を組み込んだ排水層 7 となっている。排水層 7 には砾 11 を充填する。符号 12 はネットである。

被覆土壌層 2 (厚み 5 cm) 及び浄化層 4 に於ける透水・好気性土壌 5 (厚み 5 mm) は、ゼオライト粒 (2 ~ 3 mm φ) を使用した。

一方、易透水・嫌気性土壌 6 としてはマサ土に鉄粒 (1.0 ~ 2.0 メッシュ) を 5% 混合したものを用いた。この改良された易透水・嫌気性土壌 6 の活性アルミニウム及び活性鉄の含量 (乾土重量基準) は 0.1% と 5.3% であった。そして、この易透水・嫌気性土壌 6 を  $3\text{cm} \times 5\text{cm} \times 10\text{cm}$  (—

—  $20\text{mg/l}$ ) を 1 l / 日の割合で供給した。実験は、昭和 61 年 9 月から 1 ヶ月間連続して行なった。その結果 (平均値) は、表-1 に示すように T-N、T-P とも 9.9% 以上で、極めて満足すべきものであった。尚、この装置で 1 l / 日の供給量は、2.5 l / m<sup>2</sup> / 日の流入負荷量に相当する。

次に、従来例として第 4 図に示す装置を用いて同様の汚水浄化試験を行なった結果を同じく表-1 に示す。この従来の汚水浄化装置 15 は、第 2 図の本発明装置において、易透水・嫌気性土壌 6 の代わりに黒ボク土壌 16 を充填した難透水・嫌気性土壌ブロック 17 を用いた点のみが異り、他は全く同じものである。尚、黒ボク土壌の活性アルミニウム及び活性鉄の含量 (乾土重量基準) は 5.6% と 0.6% であった。

比較例 1 は前記実施例と同様に排水管 8 を第 4 図 (イ) の状態にして、1 l / 日の原水供給を 3 ヶ月間連続して行なった。表-1 の数値は平均値であり、浄化水 (C) の T-N 濃度は経時的に上

昇し3ヶ月で浄化能が著しく低下した。

比較例2は、比較例1に引き続いて1ℓ/日の負荷水量で2ヶ月間連続して原水を供給した。但

表-1

流入水質		T-N	T-P	備考
		40 mg/ℓ	20 mg/ℓ	
上段	実施例	0.4 99.0%	0.02 99.9%	3ヶ月
放流段	比較例1	20.4 49.1%	0.16 99.2%	1ℓ/日 3ヶ月
水質除 去率 mg/ℓ % ～	比較例2	10.1 74.7%	0.16 99.2%	1ℓ/日 2ヶ月 湛水

し、この場合は排水管8を第4図の(口)の状態にして浄化層4を湛水状態(嫌気状態)にして使用した。その結果、浄化水中のT-Nが当初5mg/ℓ程度まで低下したが再び上昇し、2ヶ月後にはT-Nの浄化能が著しく低下した。尚、比較例1、比較例2とも、T-Pの除去率は99%以上

であった。

以上の比較例の結果から見て、従来型装置では浄化水(C)のT-Nの目標処理水質を10mg/ℓとした場合、長期的には流入負荷量は25ℓ/m<sup>2</sup>・日程度が限度であると思われる。

表-2

流入水質		T-N	T-P	備考
流入 負荷量		36.6 mg/ℓ	21.4 mg/ℓ	
上段	25	1.6 96.7%	0.05 99.8%	1ヶ月
放流水質 下段 mg/ℓ 除 去 率 % ～	50	1.7 99.5%	0.003 99.9%	1ヶ月
	125	2.1 94.3%	0.005 99.9%	2週間
	250	6.6 81.3%	0.18 99.2%	2週間
	375	13.4 65.2%	3.20 85.3%	2週間

次に、本発明装置がどの程度の流入負荷量に耐えるかを実験してみた。即ち、昭和62年5月から原水(A)の供給量を増やして得られた浄化水(C)の浄化の程度を調べる実験を行なった。その結果(各期間中の平均値)を表-2に示す。

尚、装置9は前記実施例と同じものを用いた。原水(A)は、人工汚水( $\text{NO}_3-\text{N}$  36.6mg/ℓ +  $\text{PO}_4-\text{P}$  21.4mg/ℓ)を用い、その供給は表中備考欄に示す通りに行いこれらを連続して計3ヶ月半行なった。また、表-2中の流入負荷量(ℓ/m<sup>2</sup>・日)は、この装置での供給量を当たりに換算したものである。この結果、本発明装置では浄化水(C)のT-Nの目標処理水質を10mg/ℓとした場合、250ℓ/m<sup>2</sup>・日程度の流入負荷量及び高速処理に十分耐え得ることが判明した。これは、施工面積やコストを勘案して実用に十分耐え得るものである。

上記実施例に示した装置は、易透水・嫌気性土壌6をジユート製袋体13に充填して土壌ブロック化して使用したものであるが、勿論これに限定

されるものではない。例えば、第3図に示すように透水・好気性土壌5と易透水・嫌気性土壌6を層状に多段化(図では2段)した汚水浄化装置18も十分な脱窒能力を発揮させることができある。

この汚水浄化装置18は、汚水吸水管1の周りに砾19を充填し浄化層4との間にネット20を配設したものである。浄化層4の上部を占める透水・好気性土壌5としては、前記したゼオライトの他、マサ土、砂等が使用できる。浄化層4の下部の易透水・嫌気性土壌6としては、マサ土や砂等に鉄粒を5%程度混入した改良土壌が使用できる。さらに、炭素源として前記炭素率(C/N比)の高い物質を混入する。その他、汚水の供給源として汚水槽や汚水栓も考えられる。

要は、本発明は透水・好気性土壌5と易透水・嫌気性土壌6の層やブロックを組み合わせて浄化層4とし、且つ易透水・嫌気性土壌6として透水性に優れた土壌に鉄粒その他の還元剤を混入したものであり、汚水浄化装置の他の部分の構成には

何ら限定するものではない。

#### 【発明の効果】

以上詳述したように、本発明の汚水浄化方法は、透水性土壌に還元剤を混入した易透水・嫌気性土壌を使用し、この土壌層に供給した汚水中の酸素を消費させることにより土壌層を強制的に嫌気状態とし、脱窒菌の活性を向上させて脱窒効果を飛躍的に向上せしめるものである。

更に、この易透水・嫌気性土壌と透水・好気性土壌とを多段に組み合わせて、透水・好気性土壌中で汚水中のSS分等の有機物の好気的分解や除去を行なわせると同時にアンモニア態窒素の硝化をはかり、易透水・嫌気性土壌中で脱窒及び脱磷させて汚水の総合的な浄化を図るものである。

従って、嫌気性土壌層の透水性向上とあいまって装置の汚水処理可能容量を大幅に増大させ、高負荷運転が可能となり、装置をコンパクト化できるため、施工場所の選定を含めて施工及びコスト面でも従来にない優れた効果を奏するものである。

又本発明の汚水浄化装置は、嫌気性土壌として入手が簡単な砂やマサ土を使用し、還元剤としても鉄粒等を用いるので、安価且つ容易に構築できるとともに、使用する土壌全体が透水性に優れており、コンパクトな装置で大量の汚水を処理することができる。また、易透水・嫌気性土壌をジュート製袋体等に充填してブロック化すると、易透水・嫌気性土壌層全体として見ると還元剤と透水性土壌との混合割合の均一化が図れるとともに、取り扱いが簡単になる利点がある。

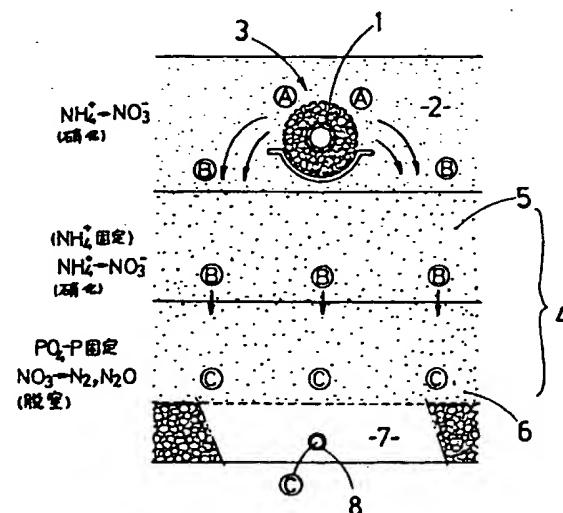
#### 4 図面の簡単な説明

第1図は本発明の汚水浄化原理を説明する概略図、第2図は本発明にかかる実験室規模の汚水浄化装置の一例を示し(a)は縦断面図、(b)は同図(a)におけるX-X線部分で断面した装置の縦断面図、第3図は実験室規模の汚水浄化装置の他の例を示し(a)は縦断面図、(b)は同図(a)におけるY-Y線部分で断面した装置の縦断面図、第4図は比較例を示し(a)は縦断面図、(b)は同図(a)におけるZ-Z線

部分で断面した装置の縦断面図である。

- 1 ……汚水吸水管
- 2 ……被覆土壌層
- 3 ……浄化層
- 4 ……透水・好気性土壌
- 5 ……易透水・嫌気性土壌
- 6 ……18汚水浄化装置
- 13 ……ジュート製袋体
- 14 ……易透水・嫌気性土壌ブロック
- A ……汚水(原水)
- B ……処理水
- C ……浄化水

第1図

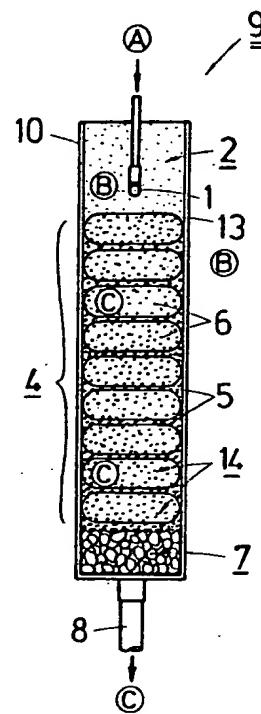
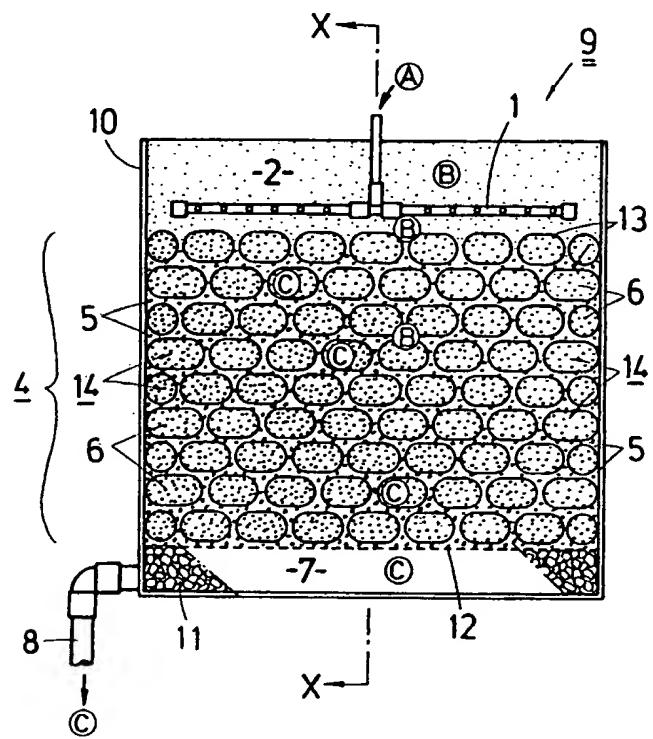


特許出願人 カナツ技建工業株式会社  
代理人 弁理士 永田久喜

## 第2回

(a)

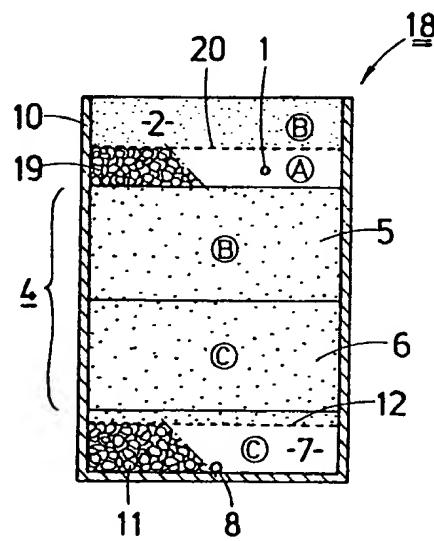
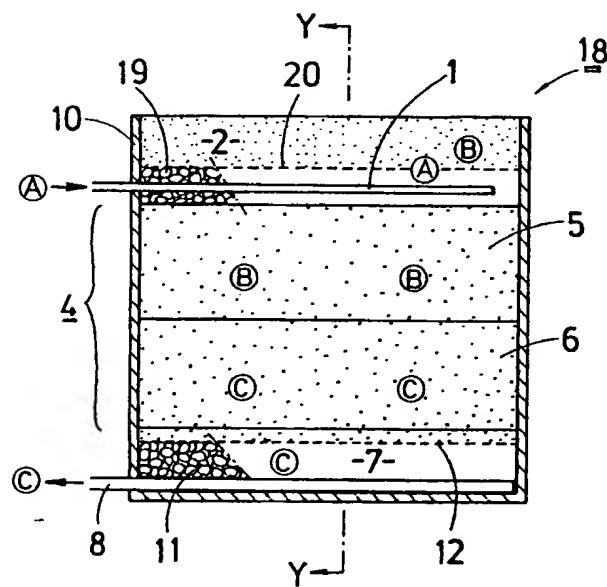
(b)



### 第3圖

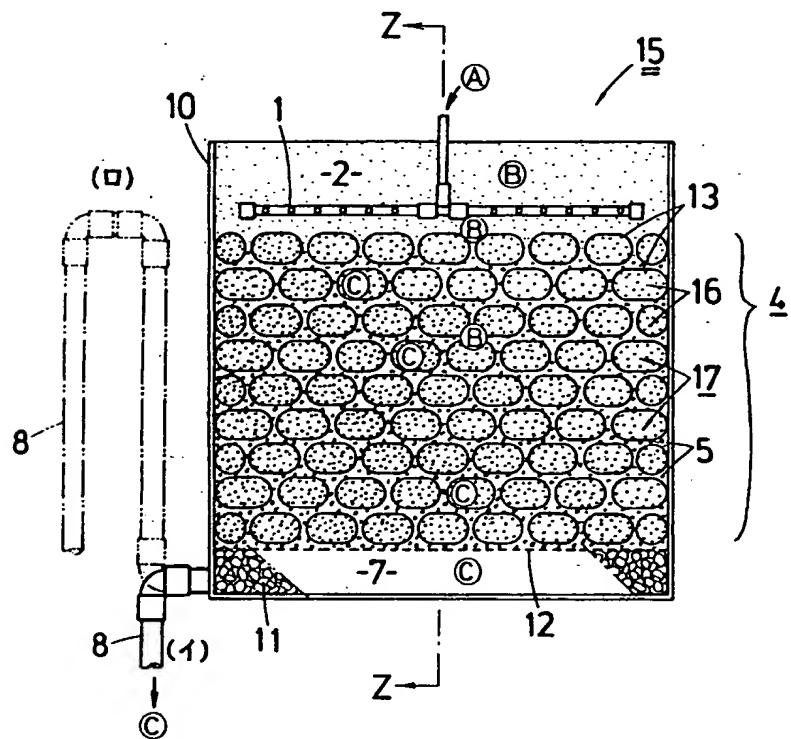
(a)

(b)

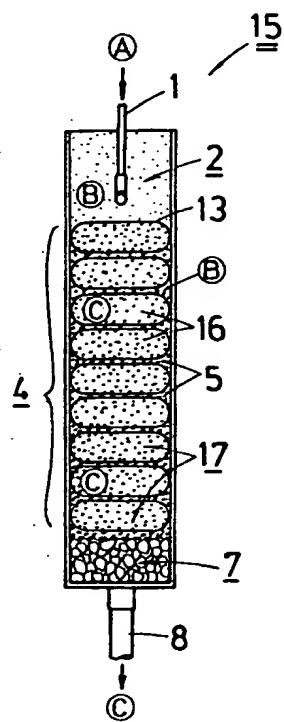


## 第4回

(a)



(b)



PURIFICATION METHOD OF POLLUTED WATER AND ITS EQUIPMENT  
[Osui no joka hoho oyobi sochi]

Toshiyoshi Wakatsuki et al

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and its equipment

## Specification

### 1. Title of Invention

Purification method of polluted water and its equipment

### 2. Scope of Patent Claims

1. The purification method of polluted water is characterized in that the soil or filled earth layer is maintained in its poor aeration state by the consumption of the oxygen in the polluted water, by supplying the polluted water into the soil or filled earth layer that is mixed with the metal iron, the denitrification process is carried out effectively by improving the activity of the denitrifying bacteria.

2. The purification method of polluted water is characterized in that the polluted water is passed through the water permeable and aerated soil or filled earth layer, the ammonia state nitrogen in the polluted water is nitrified, the denitrification process is carried out with the soil or filled earth layer mixed in with the metal iron.

3. The purification method of polluted water of Claim 1 or 2 is characterized with not only the mixing in of the metal iron but also a carbon source.

4. The soil type of purification equipment is characterized with the placement of the combination of some blocks or layers of water permeable and aerated soil or filled earth and some blocks or

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<sup>1</sup> Numbers in the margin indicate pagination in foreign text.

layers of soil or filled earth that were mixed with the metal iron, these are placed directly below the polluted water supply source.

### 3. Detailed explanation of the invention

#### [Industrial field of use]

The invention pertains to a processing method of polluted water, it pertains to a new method and the equipment used for the processing of a large quantity of polluted water using soil, the nitrogen and phosphorus that contained in polluted water such as in sewage, ammonia water, dish water, etc, exit from a household are processed.

#### [Prior Art]

The problems in the soil purification method that was offered conventionally and the properties required for the soil are outlined in the 3 points given below.

(1) The water permeation is too high, it is difficult for blockage to occur.

(2) Phosphoric acid is absorbed such as the active aluminum and iron, the content of chemical compound is high.

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(3) In order to have a good aerated soil for the nitrification of ammonia nitride, a poorly aerated soil layer must coexists so as to supply the required carbon source for the denitrification action by the organisms, the polluted water must be able to pass through at a sufficient speed and there is sufficient permeation through both layers of soil.

In order to satisfy the above requirements, the inventor

combine a soil layer that have excellent aeration and water permeation (the layer of soil is made up of sand, sandy soil, zeolite particles: this is known below as "good aeration and water permeation layer") and a soil layer that is rich in active iron, carbon source and active aluminum that have poor aeration and water permeation (black rock, red clay : known below as "difficult water permeation and poor aeration soil"). Various soil purification methods and equipment were developed using these requirements (for example, Patent Publication No. 60 - 52729, Patent Publication No. 61 - 212386).

Furthermore, as shown in figure 4, a vessel consists of a difficult water permeation and poor aeration soil layer 16 and a difficult water permeation and poor aeration soil layer are formed as the soil block 17 that are stuffed inside bag 13, the problems encountered in the implementation are resolved. Also, the material used for the vessel and bag are such as wood and jute. The denitrification capability is improved significantly by using the high carbon ratio (C/N ratio) of these materials (Patent Publication No. 61 - 10730).

(The problems resolved by the invention)

For the conventional multilayer soil purification method as described above, several problems exist in the difficult water permeation and poor aeration soil layers (the soil blocks). That is, in particular, the requirements in the water permeation and soil aeration must be satisfactory in order for the nitrification of the ammonia nitride to be successful.

In contrast to this, in the difficult water permeation and poor soil aeration, the supply of the carbon source must be sufficient for the denitrification action by the microorganisms so when considering the poor aerated soil layer, there should be high level of microorganism activity (denitrifying bacteria) and stabilization of the denitrification capability. That is, the nitrogen purification capability depends on the denitrification reaction speed of the denitrifying bacteria. The polluted water passes through the edge of the blocks and it does not pass through the difficult water permeation and the poor aeration soil layer. Therefore, when the flowing in rate ( $l/m^2 \cdot day$ ) is increased, the purification rate is reduced as the flow rate at the edge parts are increased. Also, when the flow in load is fixed, the purification capability is proportional to the thickness of the multilayer soil but as the capacity of the load ( $l/m^2 \cdot day$ ) is fixed, the thickness of the soil layer is thick (for example, twice as thick), the flow in load amount is increased (for example, twice as much), the purification rate is decreased. Therefore, the capacity ( $l/m^2 \cdot day$ ) of the processed polluted water in the equipment is increased so the surface area is increased. Thus, the implementation site is limited, the cost of production is high so the implementation of this method is limited.

[Means for resolving the problems]

The purpose of the invention is to resolve the above problems, by using the improved soil (known below as "easy water permeation and poor aerated soil") mixed with iron instead of the difficult

water permeation and poor aerated soil which have an important effect on the denitrification action, the processing of the purification is improved, the activity of the denitrifying bacteria is improved, the denitrification capability is improved significantly.

That is, when the metal iron such as the reduction iron comes into contact with the water containing air, the small amount of iron ions are eluted due to the oxidation action of the oxygen in the air in the middle region. By using this phenomenon, the soil layer is kept in the poorly aerated atmosphere by consuming the oxygen in the polluted water, the activity of the denitrifying bacteria is improved. In addition, the water permeation of the poor aeration soil region is improved, the denitrification action is improved.

The metal iron is not limited to pure iron. Also, reduction iron is preferred since it is reactive but the use is not limited to this. The form of the metal iron is preferred in the particle form since it is easy to treat and melt. The size is preferred in the range of 5 - 20 mesh.

The usage proportion of the metal iron is determined by experiment or it is calculated based on the amount of polluted water being processed, the soluble oxygen amount and the concentration of the nitrogen in the source water (the polluted water) and also the size of the iron metal and the purity of the metal iron. For the case of the iron particle content, it is preferred in the range of 4 - 6 %, in particular, it is usually in

the range of 2 - 10 wt. %. If this is less, the denitrification efficiency is reduced. If it is more, there is the problem of the elution of the iron ions.

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Furthermore, the iron ion produced from the melting of the metal iron is deposited and bond with the phosphoric acid ions, excellent effect is displayed in the removal of the phosphor.

An example of the soil that makes up the easy water permeation and poor aerated soil is such as sand, sandy soil, loamy soil, fine particles volcanic ash, coarse particles of brown petrified wood. Also, the examples of the filling material used instead of the soil used is the man made or natural particles such as zeolite particle and perlite, vermiculite, etc. In addition, pulverized plastic can be used. When the amount of the carbon content in the filling material and the soil are low, the carbon source that can be mixed in are such as jute, wheat, leaves from tree, other plants, these types of material with high carbon ratio (C/N ratio) are mixed in.

The soil with easy permeation and poor aeration are filled into the equipment, the soil is stuffed into a vessel and bag which has good water permeation property, this is a type of soil block and works extremely easily. Also, the usage amount of the soil with good aeration and easy water permeation depends on the content of the metal iron so the difference in the specific weight of the metal iron and soil is calculated. Also, the material used for the bag and vessel have high carbon rate (C/N ratio), for example, wood material and jute, therefore, the denitrification capability is

improved.

On the other hand, an example of the soil with poor aeration and easy water permeation to be filled in between the block or the aforementioned soil with easy water permeation and good aeration is such as sand and sandy soil. In addition, other types of filling materials are such as zeolite particles to be filled in the soil with poor aeration and easy water permeation.

The important role of the soil with good aeration and easy water permeation is so that the polluted water can have good contact, be dispersed and penetrates effectively in the block of soil in the layers of soil with good aeration and easy water permeation and also the equipment is not blocked up, the water permeates quickly. Also, with the soil of good aeration and easy water permeation, this acts as a means for odor removal, nitrification and decomposition of organic matter, such as the SS component, the BOD and COD components. Therefore, it is important to have good aeration in the soil and the water permeation is easy in the soil (for example, the water saturation permeation coefficient is greater than  $10^{-2} - 10^{-3}$ ). In this case, it is preferred that to promote the water permeation, sand and plants, wood of suitable size can be mixed into the soil.

When zeolite is used in the soil of good aeration and water permeation, zeolite can hold the ammonium ion well. The absorbed ammonium ion is converted into nitric acid after the action of the nitrifying bacteria, it is separated from the zeolite particle. Then, the ammonium ion is reabsorbed and the whole process is

repeated. The holding time of the nitrogen in the equipment is long so it is an effective process in the removal of the nitrogen. In addition, CEC of large zeolite is nitrified and the buffer action can be maintained against the pH reduction of the polluted water. Therefore, microorganisms in the system can be protected.

[Action]

In addition, as shown in the model in figure 1, the polluted water supplied from the polluted water distribution pipe 1 (source water) (A) used as the polluted water supply source received removal action and aeration decomposition by the organism of the SS component and the BOD and COD components due to the filtration action and the absorption by the soil and the nitrification decomposition action of the organism in the soil made up of layer 2, the sand and sandy soil. Also, it is nitrified due to the action of the nitrifying bacteria and ammonia nitride, this becomes the processed water (B).

A part of this processed water (B) is evaporated from the surface of the layer 2 but a large part is penetrated heavily into the purification layer 4 below the trench 3. The purification layer 4 is the pile up layer (it is layer 2 in the diagram) made up of the layer 6 with poor aeration and easy water permeation and layer 5 of good aeration and easy water permeation. The purification layer 4 consists of the soil layer 6 of poor aeration and easy water permeation filled into a bag made from jute, the blocks of soil of poor aeration and easy water permeation is placed with spaces on the left and right, front and back and top and

bottom, soil layer 5 of good aeration and easy water permeation are filled in these spaces.

The processed water (B) permeated into the soil layer 5 of good aeration and easy water permeation is placed under oxidation condition, this receives nitrification action and good organism decomposition similar to soil layer 2. Furthermore, when zeolite is used, ammonia state nitrogen is solidified and nitrified.

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Next, the processed water (B) is permeated into soil layer 6 of poor aeration and easy water permeation. It is placed in contact with a reduction agent such as iron, the following reaction is generated.

Fe (monomer) ...> Fe...

Thus, a large amount of the oxygen in the processed water (B) and easy permeation water and poorly aerated soil 6 are consumed. The whole layer of easy water permeation and poorly aerated soil 6 is kept in the poorly aerated state due to this action, the activity of the denitrifying bacteria is improved. Therefore, the NO<sub>2</sub> and the NO<sub>3</sub> - N (nitric acid state nitrogen) in the processed water (B) is converted to N<sub>2</sub> and N<sub>2</sub>O by the denitrifying bacteria when the middle layer of the soil layer 6 is permeated in the downflow, the denitrification is carried out effectively. Also, with this process, the phosphoric acid in the processed water (B) (positive and poly) becomes a precipitate of iron phosphate reacting with the iron ion Fe<sup>2+</sup> (+) in the easy water permeation and poorly aerated soil, this is absorbed and solidified into the

middle layer of the soil layer 6.

The nitrogen component and the phosphor that are removed largely are added to these organic matter such as the SS component, the BOD and the COD components, this purified water (C) is collected into the water exhaust layer 7, this is exhausted into the outside of the equipment via the exhaust pipe 8.

[Implementation example]

Next, the invention is explained in detail based on the implementation example shown in the diagrams.

Figure 2 show an example of the polluted water purification equipment of the laboratory model pertaining to the invention. This polluted water purification system 9 consists of various soil in the tank 10 made from acrylic of inner dimension measuring a height of 10 cm, length 45 cm and depth of 45 cm. That is, the soil layer 2 is arranged with the polluted water exhaust pipe 1 from the upper part, the middle part is the purification layer 4, the bottom part becomes the water exhaust layer 7 combined with the exhaust water pipe 8. Fill piece 11 is filled into the water exhaust layer 7. The symbol 12 is a net.

The water permeation and good aerated soil 5 (thickness 5 mm) in the purification layer 4 and the soil layer 2 (thickness of 5 cm) use the zeolite particles (2 - 3 mm diameter).

On the other hand, iron particles (10 - 20 mesh) in the sandy are used as the easy water permeation and poorly aerated soil layer 6 are mixed together at 5 %. The content of the active iron and the active aluminum of this improved easy water permeation and poorly

aerated soil layer 6 is 0.1 % and 5.3 % (the dry weight standards). Then, the easy permeation water and poorly aerated soil 6 are filled (200 g) into the jute bag 13 measuring 3 cm x 5 cm x 10 cm (one part 3 cm x 2.5 cm x 10 cm). The easy water permeation and poorly aerated soil block 14 is arranged in a space of 5 mm to the bottom and top and left and right of the above soil layer. The soil block 14 of each layer is arranged about 2.5 cm that contact and permeates the processed water (B) sufficiently. The soil block 14 that is arranged with 9 layers having a total block of 77.

This jute bag 13 is made into a unit not only filled with easy permeated water and poorly aerated soil but also can be a mesh unit that exists at the edge of the good aerated soil and poorly aerated soil. The permeation and movement of the water are possible in the direction that is at the contact edge of both layers. Also, this jute bag 13 has carbon rate (C/N ratio) that is high (normally above 50), it becomes the carbon source for the denitrifying bacteria. Furthermore, an example was given for the structure of the aforementioned polluted water purification equipment 9 and the element shape of the soil block 14 but the invention is not limited to these.

Furthermore, the polluted water from source water (A) (NO<sub>3</sub> - NH<sub>4</sub>O mg/l + PO<sub>4</sub> - P 20 mg/l) are supplied at a proportion of 1 l/day to the polluted water purification equipment 9. The experiment is performed for a continuous period of 1 month from september 1986 (Japanese year of showa 61). The result (average value) that is satisfactory is above 99 % in T-N and T-P as shown

in Table 1. Furthermore, the supply amount of 1 l/day with this equipment corresponds to the flow load amount of 2.5 l/m<sup>2</sup>.

Next, the same results were obtained by conducting a similar polluted water purification experiment using the equipment shown in figure 4, the results are shown in Table 1 of a conventional example. The conventional polluted water purification equipment 15 is filled with black rocky soil layer 16 instead of the easy water permeation and poorly aerated soil 6 used in the equipment of the invention of figure 2, the only different points are the use of difficult water permeation soil and poorly aerated soil block 17, the rest of the structure remain the same. Furthermore, the content (dry content standard) of the active iron and the active aluminum in the black rocky soil is 5.6 % and 0.6 %.

Comparison example 1 pertains to the water exhaust pipe 8 similar to that of the aforementioned implementation example, it is in the state (i) of figure 4. The source water supply of 1 l/day is supplied continuously for 3 months. The numerical value of Table 1 is the average value. The T - N concentration of the polluted water (C) is increased as time elapsed, the purification ability in 3 months is reduced significantly.

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In comparison example 2, source water is supplied continuously for a period of 2 months at a water load of 1 l/day similar to comparison example 1.

Table 1

流入水質 (1)	T - N	T - P	備考 (6)
	4.0 mg/l	2.0 mg/l	
上段 (2)	実測 0.4	0.02	3ヶ月 (1)
	基準 99.0%	99.9%	
放下 洗段 水質 (3)	実測 20.4	0.16	1ヶ月 (8)
	基準 1.4	99.2%	
一去 水質 (4)	実測 10.1	0.16	1ヶ月 (9)
	基準 2	99.2%	溢水 (9)

Keys:

1 - inflow water quality

2 - upper level water exhaust quality (mg/l), lower level removal rate (%)

3 - implementation example, 4 - comparison example 1, 5 - comparison example 2

6 - remarks

7 - 3 months

8 - 1 l/day, 3 months

9 - 1 l/day, 2 months, overflow water

That is, in this case, the water exhaust pipe 8 is in the state of (ii) of figure 4 and the purified layer 4 is used in the overflow water state (poor aerated state). As a result, T - N in the purified water is reduced to about 5 mg/l and then rise up again. After 2 months, the purification ability of T - N is reduced significantly. Furthermore, the removal rate of T - P is above 99

% for both comparison examples 1 and 2.

As observed from the results of the comparison examples, the target process water quality of T-N of the purified water (C) in the conventional equipment is 10 mg/l and over a long period of time, the in flow rate is 25 l/m<sup>2</sup> . day.

Table 2

流入水質	T-N	T-P	備考
流入水質 流入量 負荷量	36.6 mg/l	21.4 mg/l	(3)
上段 (4)	25 96.7%	0.05 99.8%	1ヶ月(5)
放流水質 下段 除害率 (%)	50 99.5%	0.003 99.9%	1ヶ月(6)
	125 94.3%	0.005 99.9%	2週間(7)
	250 81.3%	0.18 99.2%	2週間(8)
	375 65.2%	13.4 85.3%	2週間(9)

Keys:

1 - inflow water quality

2 - inflow rate

### 3 - remarks

4 - upper level water exhaust quality (ml/l), lower level removal rate (%)

5 - 1 month

6 - 1 month

7 - 2 weeks

8 - 2 weeks

9 - 2 weeks

Next, an experiment is conducted to study the inflow rate with the equipment of the invention. That is, the supply amount of source water (A) from May 1987 (Japanese year showa 62) is increased, the degree of purification of the purified water (C) obtained is investigated. The result (the average value in each period) is shown in Table 2.

Furthermore, the equipment 9 uses the same structure as the aforementioned implementation example. The source water (A) uses artificial polluted water (NO<sub>3</sub> - N36.6 mg/l + PO<sub>4</sub> - P21.4 mg/l). The supply is carried out for 3 months continuously according to the remarks shown on the Table. Also, the inflow rate (1/m<sup>2</sup>. day) in Table 2 is calculated per m<sup>2</sup> of the supply amount in this equipment. When the result show the target process quality is 10 mg/l of the T-N of the purified water (C) in the equipment of the invention, the inflow rate of 250 1/m<sup>2</sup> . day, this have good processing speed for sufficient purification. The implementation example have shown that the purification is sufficient and the implementation area and cost are improved.

The equipment shown in the above implementation example uses the soil block where the easy water permeation and poorly aerated soil layer 6 is filled in the jute bag 13 but the use is not limited to this. For example, as shown in figure 3, the water permeation and the well aerated soil 5 and the easy water permeation and poorly aerated soil layer 6 are multilayered (2 layers shown in the diagram), this displays sufficient

denitrification ability with the polluted water purification equipment 18.

This polluted water purification equipment 18 is filled with fill piece 19 surrounding the polluted water exhaust pipe 1 and net 20 is arranged with purification layer 4. The upper part of purification layer 4 becomes the water permeation and the well aerated soil 5, zeolite, sandy soil and sand can be used for this layer. The lower part of purification layer 4 becomes the easy water permeation and the poor aerated soil layer 6, sandy soil and sand can be used for this layer, 5 % of these are mixed in the soil layer. In addition, substance with high carbon rate (C/N ratio) is mixed as the carbon source. The polluted water tank and the polluted water trough becomes the supply source of the polluted water.

The most important points of the invention are that the purification layer 4 is obtained by combining the soil blocks and the layer of permeation water and well aerated soil layer 5 and the easy water permeation and poorly aerated soil layer 6 and a reduction agent is mixed in such as iron particles in the soil which have excellent water permeation, this is the easy water permeation and poorly aerated soil layer 6. However, the parts in the polluted water purification equipment is not limited to these layers.

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[Effect of Invention]

As described above, the polluted water purification method of

the invention uses the soil of easy water permeation and good aeration that were mixed with a reduction agent in the soil of good water permeation. The denitrification effect is improved significantly with the improvement of the activity of the denitrifying bacteria. The poor aeration in the soil layers is improved by the consumption of the oxygen in the polluted water that was supplied to the soil layers.

In addition, this easy water permeation and poorly aerated soil is mixed with the water permeation and good aerated soil in the soil layers, the nitrogen in the ammonia state is nitrified at the same time, the organic matter are removed and decomposed, the organic matter are such as the SS component in the polluted water in the water permeation and good aeration soil are decomposed.

Therefore, it is possible to improve the poorly aerated and water permeated soil and a large volume of polluted water can be processed so a high volume operation is possible. Since the equipment for processing is more compact, the cost of production is low and the selection of an implementation site is simplified, an excellent effect for implementing this method is obtained.

Also, the polluted water purification equipment of the invention uses sand and sandy soil as the soil with poor aeration and since iron particles are used as the reduction agent, the cost of this process is inexpensive and implementation of this process is simple, a soil layer of excellent water permeation is obtained, a large amount of polluted water can be processed with this compact system. Also, when the easy water permeation and poorly aeration

soil is arranged in a block in a jute bag, the proportion of the water permeation soil and the reduction agent can be mixed uniformly, an easy water permeation and poorly aerated soil are mixed as a single unit, the advantage here is the simplification of the purification system.

#### 4. Brief explanation of the diagrams

Figure 1 is a structure for explaining the polluted purification theory of the invention. Figure 2 is a cross section of the system showing an example of the polluted water purification equipment used in the laboratory pertaining to the invention, (a) is the longitudinal cross section and (b) is the cross section at X-X of (a). Figure 3 is a cross section of another system showing an example of the polluted water purification equipment used in laboratory pertaining to the invention, (a) is the longitudinal cross section and (b) is the cross section at Y-Y of (a). Figure 4 is a cross section of a system showing a comparison example of the polluted water purification equipment used in the laboratory pertaining to the invention, (a) is the longitudinal cross section and (b) is the cross section at Z-Z of (a).

1 - polluted water exhaust pipe

2 - soil layer

4 - purification layer

5 - water permeation and good aerated soil

6 - easy water permeation and poor aerated soil

9,18 - polluted water purification system

13 - jute bag

14 - easy water permeation and poorly aerated soil block

A - polluted water (source water)

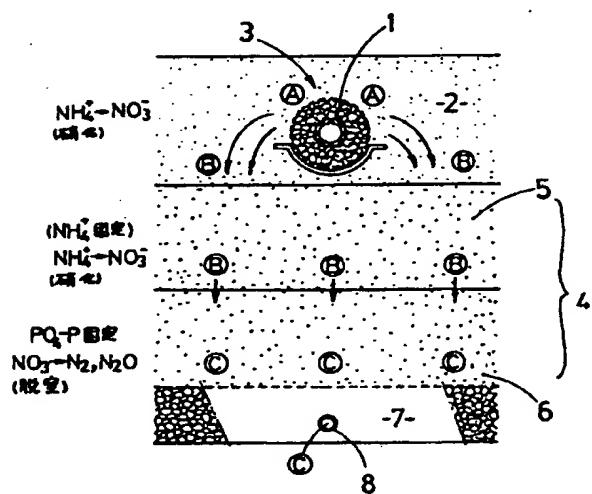
B - processed water

C - polluted water

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Figure 1



$\text{NH}_4^+ - \text{NO}_3^-$  (nitrification)

$\text{NH}_4^+$  solid

$\text{NH}_4^+ \rightarrow \text{NO}_3^-$  (nitrification)

$\text{PO}_4 - \text{P}$  solid

NO<sub>3</sub> ..> N<sub>2</sub>, N<sub>2</sub>O (denitrification)

Figure 2

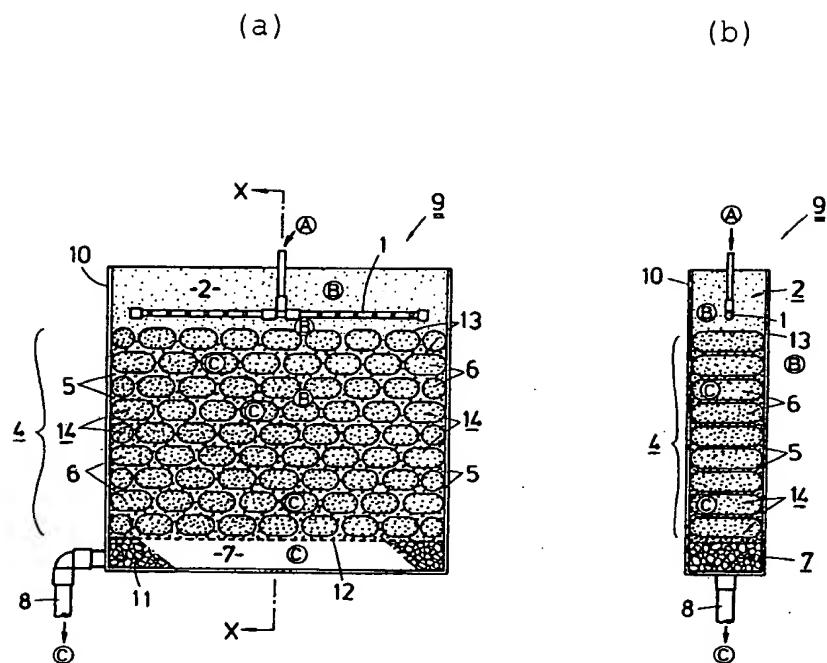


Figure 3

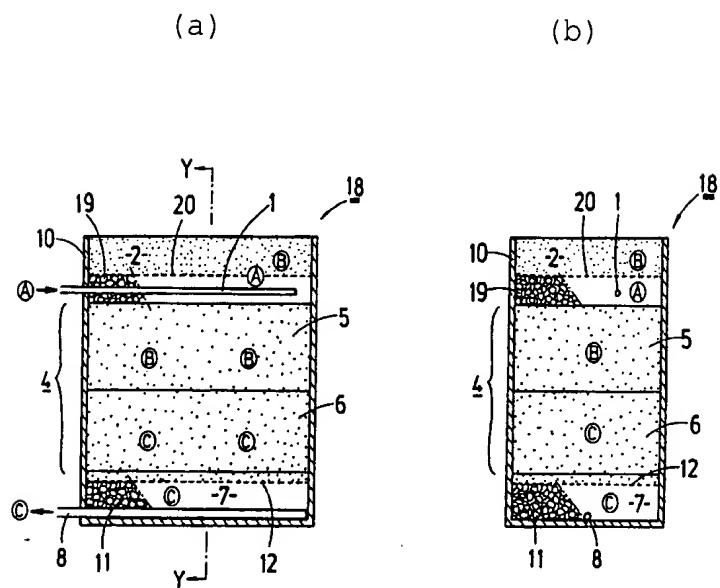


Figure 4

